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1/77

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1. Your reference HL80195/001
2. Patent application number 25 APR 2002  
(The Patent Office will fill in this part) 26APR02 E714128-1 002847  
0209535.4 P01/7700 0.00-0209535.4
3. Full name, address and postcode of the or of each applicant (*underline all surnames*)  

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837137 9001

Patents ADP number (*if you know it*)  
If the applicant is a corporate body, give the country/state of its incorporation United Kingdom
4. Title of the invention  
FLAME RETARDENT POLYMER COMPOSITIONS.
5. Full name of your agent (*if you have one*) Haseltine Lake & Co.  

Imperial House  
15-19 Kingsway  
London WC2B 6UD

Patents ADP number (*if you know it*) 34001
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Country	Priority application number ( <i>if you know it</i> )	Date of filing ( <i>day/month/year</i> )
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Claim(s) 3

Abstract 1

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11. I/We request the grant of a patent on the basis of this application

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 Date

24 April 2002

12. Name and daytime telephone number of person to contact in the United Kingdom

Mr. David Brown

[0117] 9103200

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## FLAME RETARDANT POLYMER COMPOSITIONS

### Field of the Invention

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The present invention relates to flame retardant polymer compositions, and particularly to such compositions which include particulate kaolin clays. The invention also relates to particulate filler materials for the compositions, to process intermediates from which the compositions may be formed and to articles made from the compositions.

### Background of the Invention

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Flame retardant polymer compositions are widely used, particularly in locations where there is a risk of high temperatures and/or fire, or where the consequences of burning of the polymer composition would be catastrophic. For example, the sheathing or coating of electrical cables must meet legally specified flame retardancy standards, to limit the risk of failure of electrical systems in the event of a fire and to limit the risk of a fire being started or spread as a result of overheating of the cable by the electric current. The cable sheathing or coating will be rated to withstand a specified temperature.

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Generally speaking, flame retardant polymer compositions include additives which can have one or more of the following effects on exposure of the composition to fire: (i) char promotion, in which the combusted composition forms a solid mass ("char"), which provides an insulating layer against the fire heat, inhibiting escape of volatile combustible materials from the composition and inhibiting inward diffusion of oxygen; (ii) imparting drip resistance, in which the tendency of a thermoplastic polymer to drip when heated is reduced; (iii) promotion of heat absorption, in which the additive removes heat from the system; and (iv) promotion of heat quenching, in which the additive inhibits combustion in the gas phase by interfering with the chemical reactions which spread and maintain a flame.

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Known char forming additives include phosphorus-containing compounds, boron-containing compounds and metal salts such as alkali metal salts of sulphur-

containing compounds, which can fuse and solidify at flame temperatures, thereby creating a ceramic-like or glass-like mass which structurally supports the char.

Known drip suppressing additives for thermoplastic polymers include polytetrafluoroethylene (PTFE). The PTFE is typically present at an amount of up to  
5 about 5% by weight of the total composition, and forms fibrils which stabilise the thermoplastic polymer under molten conditions. See, for example, WO-A-99/43747 and the prior publications referred to therein and in the search report thereon, the contents of which are incorporated herein by reference.

Known heat absorbing additives include metal hydroxides or hydrates such as  
10 alumina trihydrate (ATH;  $\text{Al}(\text{OH})_3$ ) or magnesium hydroxide ( $\text{Mg}(\text{OH})_2$ ). These additives are believed to work by absorbing heat to evaporate water contained in their structure.

Known heat quenching (flaming resistance) additives include free radical scavengers such as organic halogen-containing compounds such as brominated and  
15 chlorinated hydrocarbons. These additives are believed to work by releasing halogens into the flame, which inhibit combustion of the gas phase. Synergistic co-additives such as antimony oxide may be present, to enhance the heat quenching effects of the free radical scavengers. See, for example, US Patent No. 4582866 and the prior publications referred to therein and in the search report thereon, the contents of which  
20 are incorporated herein by reference.

The known additives are not entirely satisfactory, however, and the need for alternative and improved additives remains. For example, additives such as PTFE can adversely affect the surface finish of the composition. The use of halogen-containing compounds is believed to cause health problems and environmental  
25 damage. The additives can also adversely affect impact strength and impact resistance of the composition, or other physical properties. At the same time, cost pressures can urge that the level of additive used is as low as possible.

Proposals have been made to include certain clays as flame retardant additives in polymer compositions, in an attempt to answer some of these difficulties. WO-A-  
30 99/43747 and US Patent No. 4582866, referred to above, teach the inclusion of an organoclay as a co-additive.

WO-A-01/46307, the disclosure of which is incorporated herein by reference, describes polypropylene, ABS (acrylonitrile-butadiene-styrene) copolymer, polystyrene and polyurethane compositions (all thermoplastic polymers) containing as

flame retardant additive 5 or 10 parts by weight of a montmorillonite organoclay cation-exchanged with diethyl-di(hydrogenated tallow)-ammonium ion (Claytone HY), the polypropylene compositions containing either 10 parts by weight of the organoclay as sole flame retardant additive or 10 parts by weight of the organoclay together with antimony oxide and a brominated hydrocarbon selected from ethylene, bis-tetrabromo-phthalidimide and decabromodiphenyloxide. It is reported (Table 1) that the compositions all show no dripping under the Underwriters Laboratories standard 94 ("UL 94") vertical flame test (ASTM 3801), test specimens 0.062 inches (1.57 mm) thick.

US Patent No. 5946309, the disclosure of which is incorporated herein by reference, describes generally a coarse particle size kaolin clay product having an average equivalent particle diameter of about 4.5 to 6.0 microns ( $\mu\text{m}$ ) as measured using a Micromeritics Sedigraph 5100 unit, and a BET surface area of about 8 to 11  $\text{m}^2/\text{g}$ , and its use as a filler for polymeric compositions. The preferred product is stated to have a high aspect ratio, preferably of about 12 to 14 as determined by Sphericity Model calculations from experimentally determined surface area data according to the method described in US Patent No. 5167707 and the references cited therein (the contents of which are also incorporated herein by reference).

US Patent No. 5846309 specifically describes (Examples 6 and 7) a paste for making a moulded thermoset unsaturated polyester resin having a styrene content of about 33% (Aristech Resin MR 13017) containing a kaolin/ATH filler at a filler loading of 100phr (i.e. 50:50 weight percent polymer:filler). The kaolin had an equivalent particle diameter of 5.25 $\mu\text{m}$  and an aspect ratio (Sphericity Model) of 13.1 (see Table 1-C). The two ATHs used had BET surface areas of 0.24 and 2.0  $\text{m}^2/\text{g}$  (Table 6). The weight ratio of the kaolin to the ATH varied from 100:0 to 0:100 (Figures 3 and 4). The paste compositions were tested for viscosity, to determine whether the presence of the clay assisted or hindered processing of the paste. The pastes were not set and flame retardancy of the resin was therefore not tested. Indeed, it was left open whether the filler material would or would not adversely affect the physical properties of the thermoset composite (column 22, line 60 to 67). It was reported that the presence of the clay generally increased the paste viscosity, which is undesirable for processing. It was stated (column 24, lines 7 to 13) that one must carefully balance the flame retardancy plus the viscosity reduction and specific

gravity reduction benefits of ATH use against the increased cost and reduced surface finish disadvantages in a given application to achieve the best cost versus performance properties.

The present invention is based on the surprising finding that, by using a high aspect ratio particulate kaolin having an average particle diameter less than about 4  $\mu\text{m}$  in a filler component of a polymer composition, an acceptable degree of char strength can be obtained, optionally together with drip resistance, while substantially preserving general desirable physical properties of the polymer compositions.

### Brief Description of the Invention

According to the present invention in a first aspect, there is provided a flame retardant polymer composition comprising a polymer and a particulate kaolin clay having a mean equivalent particle diameter less than or equal to about 4 microns ( $\mu\text{m}$ ), e.g. less than 4.5 $\mu\text{m}$ , particularly less than 4.0 $\mu\text{m}$ , and a particle shape factor which is greater than about 30, e.g. at least about 60, particularly at least about 70, particularly at least about 90, most particularly at least about 100, e.g. at least about 120, and preferably up to about 150.

The composition may suitably include one or more further non-kaolin component, which may be selected from one or more conventional flame retardant component, one or more conventional non-flame retardant component, or both. Any non-kaolin component will suitably be present in a smaller weight proportion than the essential components of the composition. The essential components of the composition preferably constitute the majority (i.e. over half) of the weight of the composition.

The conventional flame retardant component, when present, may, for example, be selected from phosphorus-containing compounds, boron-containing compounds, metal salts, metal hydroxides, metal oxides, hydrates thereof, organoclays (including ion-exchanged and any other modified organoclays), halogenated hydrocarbons, and any combination thereof, typically boric acid, a metal borate and any combination thereof. A preferred flame retardant component is ATH.

The conventional non-flame retardant component, when present, may, for example, be selected from pigments, colorants, anti-degradants, anti-oxidants, impact

modifiers, inert fillers, slip agents, antistatic agents, mineral oils, stabilisers, flow enhancers, mould release agents, nucleating agents, clarifying agents, and any combination thereof.

According to the present invention in a second aspect, there is provided a particulate filler material for a flame retardant polymer composition, the filler material comprising a mixture of a particulate flame retardant (for example, ATH) and a particulate kaolin clay, wherein the particulate kaolin clay has a mean equivalent particle diameter less than or equal to about 4 microns ( $\mu\text{m}$ ) and a particle shape factor which is greater than about 30. The particulate filler material may further comprise one or more additional non-kaolin flame retardant component and/or one or more non-kaolin non-flame retardant component.

For processing to form the polymer composition, the components will preferably be mixed, the polymer component being present as liquid or particulate solid, optionally as one or more precursor(s) of the polymer component. Such a process and the resultant mixture constitute respectively third and fourth aspects of the present invention.

According to the present invention in a fifth aspect, there is provided an article, for example an electrical product or other article comprising a sheath, coating or housing, formed from a flame retardant polymer composition according to the first aspect of the present invention.

### **Detailed Description of the Invention**

#### *Particulate Kaolin*

The particulate kaolin is preferably a hydrous kaolin such as uncalcined kaolin or ball clay. Mixtures of different kaolins may be used, provided that the particulate kaolin has the required mean equivalent particle diameter and the required shape factor.

A kaolin product of high shape factor is considered to be more "platey" than a kaolin product of low shape factor. "Shape factor" as used herein is a measure of an average value (on a weight average basis) of the ratio of mean particle diameter to particle thickness for a population of particles of varying size and shape as measured



using the electrical conductivity method and apparatus described in GB-A-2240398/US-A-5128606/EP-A-0528078 and using the equations derived in these patent specifications. "Mean particle diameter" is defined as the diameter of a circle which has the same area as the largest face of the particle. In the measurement method described in EP-A-0528078 the electrical conductivity of a fully dispersed aqueous suspension of the particles under test is caused to flow through an elongated tube. Measurements of the electrical conductivity are taken between (a) a pair of electrodes separated from one another along the longitudinal axis of the tube, and (b) a pair of electrodes separated from one another across the transverse width of the tube, and using the difference between the two conductivity measurements the shape factor of the particulate material under test is determined.

The "aspect ratio" parameter of the kaolin clay product of the prior art US Patent No. 5946309 is not numerically the same as the "shape factor" parameter of the kaolin used in the present invention. For example, for one clay which we have tested, it is found experimentally that an "aspect ratio" of 9 according to the prior art determination corresponds to a "shape factor" according to the present invention of about  $65 \pm 5$ . Therefore, it is believed that a particulate kaolin having an "aspect ratio" of greater than 9 according to the prior art determination will probably fulfil the requirement of "shape factor" according to the present invention. However, since the average equivalent particle diameter of the kaolin used in the present invention is clearly different from that of the kaolin used in the prior art patent, the determination methods for this parameter being the same as between the prior art patent and the present invention, the products are different and an attempt to correlate aspect ratio with shape factor between such different materials has not been made.

The mean (average) equivalent particle diameter ( $d_{50}$  value) and other particle size properties referred to herein for the particulate kaolin are as measured in a well known manner by sedimentation of the particulate material in a fully dispersed condition in an aqueous medium using a Sedigraph 5100 machine as supplied by Micromeritics Instruments Corporation, Norcross, Georgia, USA (telephone: +1 770 662 3620; web-site: [www.micromeritics.com](http://www.micromeritics.com)), referred to herein as a "Micromeritics Sedigraph 5100 unit". Such a machine provides measurements and a plot of the cumulative percentage by weight of particles having a size, referred to in the art as the 'equivalent spherical diameter' (esd), less than given esd values. The mean particle size  $d_{50}$  is the value determined in this way of the particle esd at which there are 50%

by weight of the particles which have an equivalent spherical diameter less than that  $d_{50}$  value.

The value of  $d_{50}$  for the particulate kaolin is less than or equal to about  $4\mu\text{m}$ . It may, for example, be in the range  $0.4\mu\text{m}$  to about  $3\mu\text{m}$ , especially  $0.5\mu\text{m}$  to about  $2\mu\text{m}$ . For example, particulate kaolin of English (Cornish) origin may have a  $d_{50}$  value of from  $0.5\mu\text{m}$  to  $1.5\mu\text{m}$ .

The particulate kaolin according to the invention may be prepared by light comminution, e.g. grinding or milling, of a coarse kaolin to give suitable delamination thereof. The comminution may be carried out by use of beads or granules of a plastics, e.g. nylon, grinding or milling aid. The coarse kaolin may be refined to remove impurities and improve physical properties using well known procedures. The kaolin may be treated by a known particle size classification procedure, e.g. screening and/or centrifuging, to obtain particles having a desired  $d_{50}$  value.

A range of particulate kaolins are available, which have the required particle size and shape factor, or can easily be processed in ways well known to the skilled worker to arrive at the required particle size and shape factor. One suitable particulate kaolin for use in the present invention has a mean equivalent particle diameter of about  $1.3\mu\text{m}$  and a shape factor in the range of about 120 to about 150. It typically also has a specific gravity of about  $2.6\text{ g/cm}^3$ , a specific surface area of about  $11\text{ m}^2/\text{g}$  as measured by the BET nitrogen absorption method, a brightness (ISO) of about 89, a chemical analysis (by X-ray fluorescence) of 46.4%  $\text{SiO}_2$  and 38.4%  $\text{Al}_2\text{O}_3$ , and a particle size distribution such that a maximum of 3% by weight of the particles have a size greater than  $10\mu\text{m}$  and a minimum of 67% by weight of the particles have a size less than  $2\mu\text{m}$ .

The kaolin clay is suitably present in the polymer composition according to the present invention at amounts in the general loading range between about 10 and about 150 parts by weight per hundred of polymer, and more preferably between about 10 and about 100 parts per hundred.

### *Polymer*

The polymer comprises any natural or synthetic polymer or mixture thereof. The polymer may, for example, be thermoplastic or thermoset. The term "polymer"

used herein includes homopolymers and copolymers, as well as crosslinked and/or entangled polymers and elastomers such as natural or synthetic rubbers and mixtures thereof. Specific examples of suitable polymers include, but are not limited to, polyolefins of any density such as polyethylene and polypropylene, polycarbonate, polystyrene, polyester, acrylonitrile-butadiene-styrene copolymer, nylons, polyurethane, ethylene-vinylacetate polymers, and any mixture thereof, whether cross-linked or un-cross-linked.

The term "precursor" as applied to the polymer component will be readily understood by one of ordinary skill in the art. For example, suitable precursors may include one or more of: monomers, cross-linking agents, curing systems comprising cross-linking agents and promoters, or any combination thereof. Where according to the invention the particulate kaolin is mixed with precursors of the polymer, the polymer composition will subsequently be formed by curing and/or polymerising the precursor components to form the desired polymer.

#### *Flame Retarding Component*

As stated above, the polymer composition according to the present invention may suitably contain one or more non-kaolin flame retarding additives. Such additives may, for example, be selected from one or more of the following:

- (i) One or more char promoter;
- (ii) One or more drip suppressant;
- (iii) One or more heat absorber; and
- (iv) One or more heat quencher (ignition suppressant).

Any conventional such additives may be used, as will be apparent to one of ordinary skill in this art. Examples of such additives include:

#### Char promoters and drip suppressants

Phosphorus-containing compounds (e.g. organophosphates or phosphorus pentoxide), boron-containing compounds (e.g. boric acid and metal borates such as sodium borate, lithium metaborate, sodium tetraborate or zinc borate), organoclays

(e.g. smectite clays such as bentonite, montmorillonite, hectorite, saponite and ion-exchanged forms thereof, suitably ion-exchanged forms incorporating cations selected from quaternary ammonium and alkylimidazolium ions), metal oxides (e.g. lead dioxide);

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#### Heat absorbers

Metal salts, metal hydroxides (e.g. ATH, magnesium hydroxide), hydrates thereof (e.g. sodium tetraborate decahydrate);

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#### Heat quenchers

Halogenated hydrocarbons (e.g. halogenated carbonate oligomers, halogenated phenyl oxides, halogenated alkylene-bis-phthalidimides and halogenated diglycyl ethers), optionally together with metal oxides (e.g. antimony oxide).

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The non-kaolin flame retarding component, when present, is suitably present in the polymer composition or the filler material according to the present invention at amounts between about 5 and about 70% by total weight of the kaolin and non-kaolin flame retarding components, and more preferably between about 5 and about 50% by weight.

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#### *Non-Flame Retarding Component*

The polymer composition may include one or more non-kaolin non-flame retardant additives for polymers, for example selected from pigments, colorants, anti-degradants, anti-oxidants, impact modifiers (e.g. core-shell graft copolymers), fillers (e.g. talc, mica, wollastonite, glass or a mixture thereof), slip agents (e.g. erucamide, oleamide, linoleamide or steramide), antistatic agents, mineral oils, stabilisers, flow enhancers, mould release agents (e.g. metal stearates such as calcium stearate and magnesium stearate), nucleating agents, clarifying agents, and any combination thereof.

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The non-kaolin non-flame retarding component, when present, is suitably present in the polymer composition or the filler material according to the present

invention at amounts up to about 50% by total weight of the kaolin and, if present, non-kaolin flame retarding component, and more preferably between up to about 30% by weight.

### 5            *Preparation of the Compositions*

Preparation of the polymer compositions of the present invention can be accomplished by any suitable mixing method known in the art, as will be readily apparent to one of ordinary skill in the art.

10            Such methods include dry blending of the individual components or precursors thereof and subsequent processing in conventional manner.

In the case of thermoplastic polymer compositions, such processing may comprise melt mixing, either directly in an extruder for making an article from the composition, or pre-mixing in a separate mixing apparatus such as a Banbury mixer.

15            Dry blends of the individual components can alternatively be directly injection moulded without pre-melt mixing.

The filler material according to the second aspect of the present invention can be prepared by mixing of the components thereof intimately together. The said filler material is then suitably dry blended with the polymer and any desired additional  
20            components, before processing as described above.

For the preparation of cross-linked or cured polymer compositions, the blend of uncured components or their precursors, and, if desired, the kaolin and any desired non-kaolin component(s), will be contacted under suitable conditions of heat, pressure and/or light with an effective amount of any suitable cross-linking agent or curing  
25            system, according to the nature and amount of the polymer used, in order to cross-link and/or cure the polymer.

For the preparation of polymer compositions where the kaolin and any desired non-kaolin component(s) are present *in situ* at the time of polymerisation, the blend of monomer(s) and any desired other polymer precursors, kaolin and any non-kaolin  
30            component(s) will be contacted under suitable conditions of heat, pressure and/or light, according to the nature and amount of the monomer(s) used, in order to polymerise the monomer(s) with the kaolin and any desired non-kaolin component(s) *in situ*.

## *Articles*

The polymer compositions can be processed to form, or to be incorporated in, articles of commerce in any suitable way. Such processing may include compression  
 5 moulding, injection moulding, gas-assisted injection moulding, calendaring, vacuum forming, thermoforming, extrusion, blow moulding, drawing, spinning, film forming, laminating or any combination thereof. Any suitable apparatus may be used, as will be apparent to one of ordinary skill in this art.

The articles which may be formed from the compositions are many and  
 10 various. Examples include sheaths for electrical cables, electrical cables coated or sheathed with the polymer composition, and housings and plastics components for electrical appliances (e.g. computers, monitors, printers, photocopiers, keyboards, pagers, telephones, mobile phones, hand-held computers, network interfaces, plenums and televisions).

### **Brief Description of the Drawings**

Embodiments of the present invention will now be described, purely by way of example and without limitation, with reference to the later Examples and with  
 20 reference to the accompanying drawings, in which:

Figure 1 shows graphs of shear viscosity on a logarithmic vertical axis (Pa.s) plotted against shear rate on a logarithmic horizontal axis ( $s^{-1}$ ), for (a) two polymer compositions according to the present invention and (b) two control compositions not including any mineral filler; and

Figure 2 shows a graph of shear viscosity on a logarithmic vertical axis (Pa.s) plotted against shear rate on a logarithmic horizontal axis ( $s^{-1}$ ), for two further polymer compositions according to the present invention, as well as the same  
 30 compositions as shown in Figure 1(b).

### **Detailed Description of the Drawings**

#### *Preparation of Test Materials*

The following Examples illustrate the preparation of the test materials  
 35 embodying the present invention and the comparison and control materials.

"Platey" Clay

A powdered platey kaolin clay (designated Clay A) was used in the Examples. Clay A had a mean equivalent particle diameter of about  $1.3\mu\text{m}$ ; a shape factor in the range of about 120 to about 150; a specific gravity of about  $2.6\text{ g/cm}^3$ ; a specific surface area of about  $11\text{ m}^2/\text{g}$  as measured by the BET nitrogen absorption method; a brightness (ISO) of about 89; a chemical analysis (by X-ray fluorescence) of 46.4%  $\text{SiO}_2$  and 38.4%  $\text{Al}_2\text{O}_3$ ; and a particle size distribution such that a maximum of 3% by weight of the particles have a size greater than  $10\mu\text{m}$  and a minimum of 67% by weight of the particles have a size less than  $2\mu\text{m}$ .

Examples 1 to 4

The materials used for Figure 1(a) and included also in Figure 2 were prepared by compounding the following thermoplastic polymers with Clay A at a loading of 61% clay by total weight of the composition: Example 1 used Escorene UL0019, an ethylene-vinylacetate copolymer available from Exxon Corporation, and the composition also contained 2% by weight of AC400, which is an ethylene-vinylacetate co-polymer (available from Honeywell), as a plasticiser; Example 2 used Clearflex Linear Low Density Polyethylene (CLDO), available from Polimeri Europa, and the composition also contained 2% by weight of AC6, which is a polyethylene homopolymer (available from Honeywell), as a plasticiser. A conventional Brabender mixer was used for the compounding.

The composition of Example 3, one of the further compositions according to the invention included in Figure 2, was prepared by compounding Escorene UL0019 with a 50:50 by weight mixture of powdered ATH and Clay A at a total filler loading of 61% filler by total weight of the composition. A conventional Brabender mixer was used for the compounding.

The composition of Example 4, the final composition according to the invention included in Figure 2, was prepared by compounding CLDO with a 50:50 by weight mixture of powdered ATH and Clay A at a total filler loading of 61% filler by

total weight of the composition. A conventional Brabender mixer was used for the compounding.

The control materials used for Figure 1(b) and included also in Figure 2 were the unfilled Escorene UL0019 and Clearflex polymers each containing 2% of the respective plasticiser. A conventional Banbury mixer was used for the compounding.

#### **Comparative Examples C1 and C2 and Examples 5 to 11**

Example 3 was repeated, but replacing the following proportions of ATH:Clay A for the 50:50 ratio previously described.

Comparative Example C1: Escorene UL0019 + 2% AC400 + 61% ATH;

Example 5: Escorene UL0019 + 2% AC400 + 61% filler (90:10 by weight ATH:Clay A);

Example 6: Escorene UL0019 + 2% AC400 + 61% filler (70:30 by weight ATH:Clay A);

Example 7: Escorene UL0019 + 2% AC400 + 61% filler (60:40 by weight ATH:Clay A);

Example 8: Escorene UL0019 + 2% AC400 + 61% filler (40:60 by weight ATH:Clay A);

Example 9: Escorene UL0019 + 2% AC400 + 61% filler (30:70 by weight ATH:Clay A);

Example 10: Escorene UL0019 + 2% AC400 + 61% filler (50:30:20 by weight Clay A:ATH:zinc borate);

Example 11: Escorene UL0019 + 2% AC400 + 61% filler (30:70 by weight zinc borate:Clay A);

Comparative Example C2: Escorene UL0019 + 2% AC400 + 61% filler (5:95 by weight Claytone™ AF organoclay (from Southern Clay Products):ATH).

#### **Comparative Examples C3 and C4 and Examples 12 to 18**

Example 4 was repeated, but replacing the following proportions of ATH:Clay A for the 50:50 ratio previously described.



Comparative Example C3: CLDO + 2% AC400 + 61% ATH;

Example 12: CLDO + 2% AC400 + 61% filler (90:10 by weight ATH:Clay A);

Example 13: CLDO + 2% AC400 + 61% filler (70:30 by weight ATH:Clay A);

5 Example 14: CLDO + 2% AC400 + 61% filler (60:40 by weight ATH:Clay A);

Example 15: CLDO + 2% AC400 + 61% filler (40:60 by weight ATH:Clay A);

Example 16: CLDO + 2% AC400 + 61% filler (30:70 by weight ATH:Clay A);

Example 17: CLDO + 2% AC400 + 61% filler (50:30:20 by weight Clay A:ATH:  
zinc borate);

10 Example 18: CLDO + 2% AC400 + 61% filler (30:70 by weight zinc borate:Clay A);

Comparative Example C4: CLDO + 2% AC400 + 61% filler (5:95 by weight  
Claytone™ AF organoclay:ATH).

### *Test Methods*

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#### Viscosity measurements

Viscosity measurements of the polymer compositions of Examples 1 to 4 and  
the controls were carried out using a Rosand capillary extrusion rheometer at 130°C  
20 and speeds sequence of 200, 50, 20, 10, 5, 2, 1, 0.5, 1, 2, 5, 10, 20, and 50. The  
results are shown in Figures 1 and 2 of the drawings.

#### Char Strength Measurements

25 Qualitative assessments of char strength and form were made of the polymer  
compositions of Examples 1 to 18, and Comparative Examples C1 and C3, after  
completion of the flammability test (see below). The results are shown in Tables 2  
and 3.

#### 30 Underwriters Laboratories Standard UL94 Flammability Test (ASTM 3801)

The UL94 flammability test protocol was performed on 150 x 10 x 1mm test  
samples of the polymer compositions of Examples 1 to 18, and Comparative  
Examples C1 and C3.

According to this test protocol, the test samples were clamped in a vertical position. The lower end was positioned 300mm above a cotton wool pad and ignited with a Bunsen burner blue flame of 20mm height. The flame was applied for 10sec and the burning properties were recorded and reported in Tables 2 and 3 below

5 (columns headed "Flame time to clamp" (the time taken in seconds, for the flame to reach the clamp); "Flame Dripping" (whether the polymer composition dripped during burning); "Cotton Ignition" (whether the cotton wool pad was ignited by any dripping polymer); "Char Strength" (a visual assessment of the nature and strength of any char); "V rating" (a flammability rating according to the test method; the assigned

10 V rating in Tables 2 and 3 is not authoritative, as the test sample dimensions were smaller than the prescribed dimensions in the standard test (13mm width)). The results are shown in Tables 2 and 3.

#### Oxygen Index

15 The oxygen index test was carried out on 70 x 4 x 2mm test samples of the polymer compositions of Examples 1 to 4, as well as Comparative Examples C1 and C3. The test used an oxygen index machine, which measured the minimum concentration of oxygen in a flowing mixture of oxygen and nitrogen that just

20 supported flaming combustion of the burning polymer. The test samples were clamped in a vertical position inside the glass chimney of the machine and ignited and burnt from top downward. The oxygen index (OI) is expressed in terms of this oxygen concentration and values for the above compositions are reported in Tables 2 and 3.

#### Tensile Strength

25 The tensile strength of the polymer compositions was measured in conventional manner. The data (expressed in MPa) are shown in Tables 2 and 3.

#### Elongation

30 The percentage elongation at breaking was measured in conventional manner on the polymer compositions. The results are shown in Tables 2 and 3.

**Table 2**

Fire properties							Mechanical Properties	
Composition	Flame time to the clamp (sec)	Flame Dripping	Cotton ignition	Char Strength	V rating	Oxygen Index (OI)	Tensile strength MPa	Elongation (%)
C1	104	Yes	Yes	Ash	2	29	11	86
Example 5	120	Yes	Yes	Soft shell	2	N/a	12.59	82.8
Example 6	130	Yes	Yes	Soft shell	2	N/a	13.41	47.16
Example 7	100	Yes	Yes	Soft shell	2	N/a	13.60	35.44
Example 3	115	Yes	Yes	Soft shell	2	27	14	32
Example 8	90	Yes	Yes	Soft shell	2	N/a	16.03	22.74
Example 9	100	Yes	Yes	Soft shell	2	N/a	14.72	20.80
Example 1	112	NO	NO	Soft shell	2	23	14	13
Example 10	110	Yes	Yes	Soft shell	2	N/a	12.94	28.43
Example 11	N/a	N/a	N/a	N/a	N/a	N/a	14.18	29.95
C2	N/a	N/a	N/a	N/a	N/a	N/a	9.39	66.6

**Table 3**

Fire properties							Mechanical Properties	
Composition	Flame time to the clamp (sec)	Flame Dripping	Cotton ignition	Char Strength	V rating	Oxygen Index (OI)	Tensile strength MPa	Elongation (%)
C3	148	Yes	Yes	Ash	2	29	9	94
Example 12	128	Yes	Yes	Soft shell	2	N/a	9.81	77
Example 13	154	Yes	Yes	Soft shell	2	N/a	9.8	37.1
Example 14	125	Yes	Yes	Soft shell	2	N/a	9.48	31.89
Example 4	135	NO	NO	Soft shell	2	25	10	27
Example 15	120	NO	NO	Soft shell	2	N/a	9.08	17.50
Example 16	120	NO	NO	Soft shell	2	N/a	9.76	25.32
Example 2	133	NO	NO	Soft shell	2	23	8	23
Example 17	120	Yes	Yes	Soft shell	2	N/a	9.47	24.68
Example 18	90	Yes	Yes	N/a	2	N/a	9.03	15.60
C4	N/a	N/a	N/a	N/a	N/a	N/a	5.82	31.63

## Discussion

Referring to the results shown in Figures 1 and 2, there is little difference in the viscosity of all the compositions. The graph lines for the CLDO compositions are below the graph lines for the Eşcorene compositions, showing that the CLDO compositions have generally lower viscosity than the Eşcorene compositions. This is in agreement with the viscosity of the base polymers, as the CLDO has lower viscosity than the EVA polymer.

Viscosity measurements of the polymers composition with 61% clay and 2% plasticiser are given in Figure 1(a). Again, there is little if any difference in the viscosity of the composition as a result of inclusion of Clay A. This indicates that the ATH could be replaced in a large percentage with the clay without affecting the production speed of the polymer composition, e.g. in an electrical cable manufacturing process.

The viscosity of the compositions with 50ATH: 50 Clay A with total filler loading of 61% was also measured and the data are given in Figure 2 of the drawings. It can be concluded that there is little adverse effect on viscosity by partially substituting ATH with Clay A.

All the compositions of the Examples (i.e. according to the present invention) produced a char in the form of a shell, a significant improvement on the ash produced when ATH alone was used as filler.

Indeed, as shown in Tables 2 and 3, it is advantageous for the clay to be present with ATH. The clay may suitably be present in an amount greater than the ATH. At a clay loading equal to and above 50:50 Clay A:ATH, the clay/ATH filler stopped dripping of the molten CLDO polymer. 100% clay was required before dripping of the Eşcorene polymer was stopped. The incorporation of relatively large amounts of clay into the filler in partial substitution for the ATH does not significantly impair the other fire and mechanical properties of the polymer compositions, compared with the polymer filled with ATH alone.

Comparative Examples C2 and C4 used a mixture of Claytone AF organoclay and ATH (5:95). This is a clay of the type described in WO-A-01/46307. The clay compounded well with base polymers and the mechanical properties are given in Tables 2 and 3. While the elongation of these compositions was quite high, the tensile

strength was significantly poorer than the compositions of the present invention, and poorer even than the comparison compositions filled with ATH alone.

### *Conclusions*

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The use of a particulate kaolin in accordance with the present invention, as a flame retardant component in polymer compositions, in effective amounts and optionally in the presence of co-additives, offers significant cost and technical advantages in the formulation of flame retardant polymer compositions having generally acceptable char strength, optionally together with good drip resistance.

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The present invention has been described broadly and without limitation to specific embodiments. Variations and modifications as will be readily apparent to those of ordinary skill in this art are intended to be included within the scope of this application and subsequent patent(s).

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**CLAIMS**

1. A flame retardant polymer composition comprising a polymer and a particulate kaolin clay having a mean equivalent particle diameter less than or equal to about 4  $\mu\text{m}$  and a particle shape factor which is greater than about 30.
2. A composition according to claim 1, wherein the mean equivalent particle diameter is less than or equal to 4.0 $\mu\text{m}$ .
3. A composition according to claim 1 or 2, wherein the mean equivalent particle diameter is less than or equal to about 3 $\mu\text{m}$ .
4. A composition according to any one of the preceding claims, wherein the shape factor is greater than about 70.
5. A composition according to any one of claims 1 to 3, wherein the shape factor is greater than about 90.
6. A composition according to any one of claims 1 to 3, wherein the shape factor is greater than about 100.
7. A composition according to any one of claims 1 to 3, wherein the shape factor is greater than about 120.
8. A composition according to any one of the preceding claims, wherein the shape factor is up to about 150.
9. A composition according to any one of the preceding claims, including one or more non-kaolin flame retardant component.
10. A composition according to claim 9, wherein the flame retardant component is selected from phosphorus-containing compounds, boron-containing compounds, metal salts, metal hydroxides, metal oxides, hydrates thereof, organoclays (including

ion-exchanged and any other modified organoclays), halogenated hydrocarbons, and any combination thereof.

5 11. A composition according to claim 9, wherein the flame retardant component comprises alumina trihydrate (ATH), boric acid, a metal borate or a combination thereof.

10 12. A composition according to claim 9, 10 or 11, wherein the particulate kaolin clay is present in an amount of at least about 50% of the total weight of particulate kaolin clay and flame retardant component.

13. A composition according to any one of the preceding claims, wherein the polymer comprises a thermoplastic polymer.

15 14. A composition according to any one of claims 1 to 12, wherein the polymer comprises a thermoset polymer.

20 15. A composition according to any one of claims 1 to 12, wherein the polymer is selected from polyolefins, polycarbonate, polystyrene, polyester, acrylonitrile-butadiene-styrene copolymer, nylons, polyurethane, ethylene-vinylacetate polymers, and any mixture thereof.

25 16. A composition according to claim 15, wherein the polyolefin comprises polyethylene or polypropylene.

30 17. A particulate filler material for a flame retardant polymer composition, the filler material comprising a particulate non-kaolin flame retardant and a particulate kaolin clay, wherein the particulate kaolin clay has a mean equivalent particle diameter less than or equal to about 4  $\mu\text{m}$  and a particle shape factor which is greater than about 30.

18. A particulate filler material according to claim 17, wherein the non-kaolin flame retardant component is selected from phosphorus-containing compounds,

boron-containing compounds, metal salts, metal hydroxides, metal oxides, hydrates thereof, organoclays (including ion-exchanged and any other modified organoclays), halogenated hydrocarbons, and any combination thereof.

5 19. A particulate filler material according to claim 17 or 18, which consists essentially of ATH and the particulate kaolin, and optionally one or more other non-kaolin flame retardant component, with less than about 10% by weight of other component(s).

10 20. A process for forming the polymer composition as claimed in any one of claims 1 to 16, which comprises mixing the components of the composition, the polymer component being present as liquid or particulate solid, optionally as one or more precursor(s) of the polymer component.

15 21. A mixture of a particulate filler material as claimed in any one of claims 17 to 19 and a polymer or a precursor thereof in the form of a liquid or particulate solid.

22. An article formed from a flame retardant polymer composition as claimed in any one of claims 1 to 16.

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23. A sheath, coating or housing for an electrical product, formed from a polymer composition as claimed in any one of claims 1 to 16.

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ABSTRACTFLAME RETARDANT POLYMER COMPOSITIONS

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A flame retardant polymer composition having acceptable char strength and optionally also drip resistance comprises a polymer and a particulate kaolin clay having a mean equivalent particle diameter less than or equal to about 4  $\mu\text{m}$  and a particle shape factor which is greater than about 30, and preferably also alumina trihydrate (ATH) and/or another flame retardant.

10

Figure 1(a)

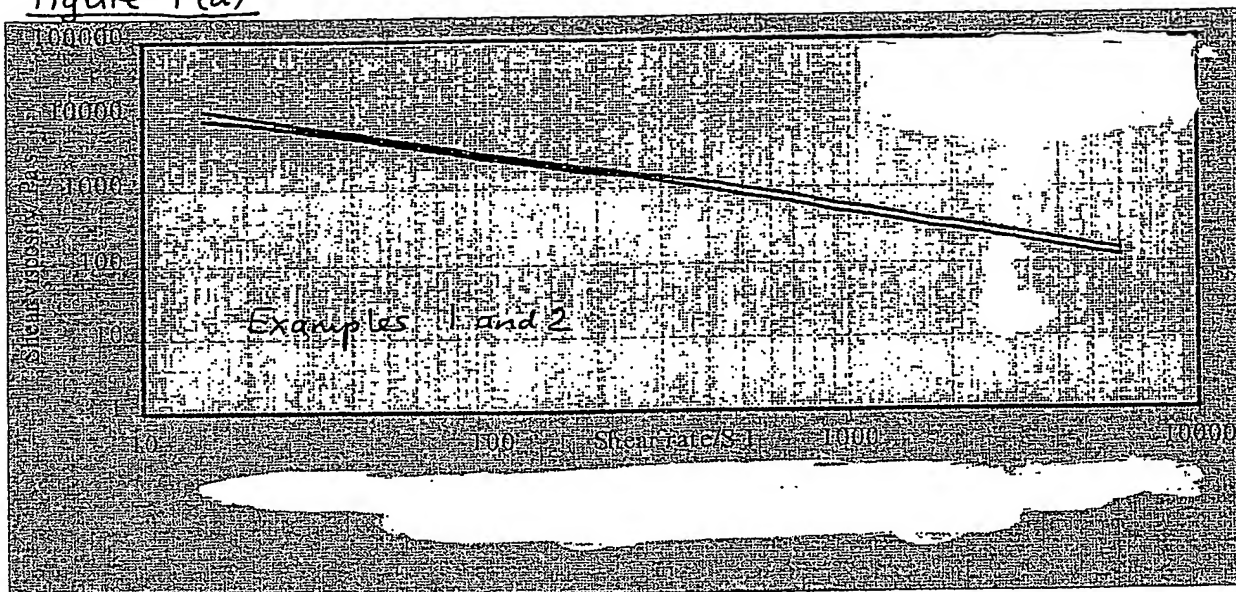


Figure 1(b)

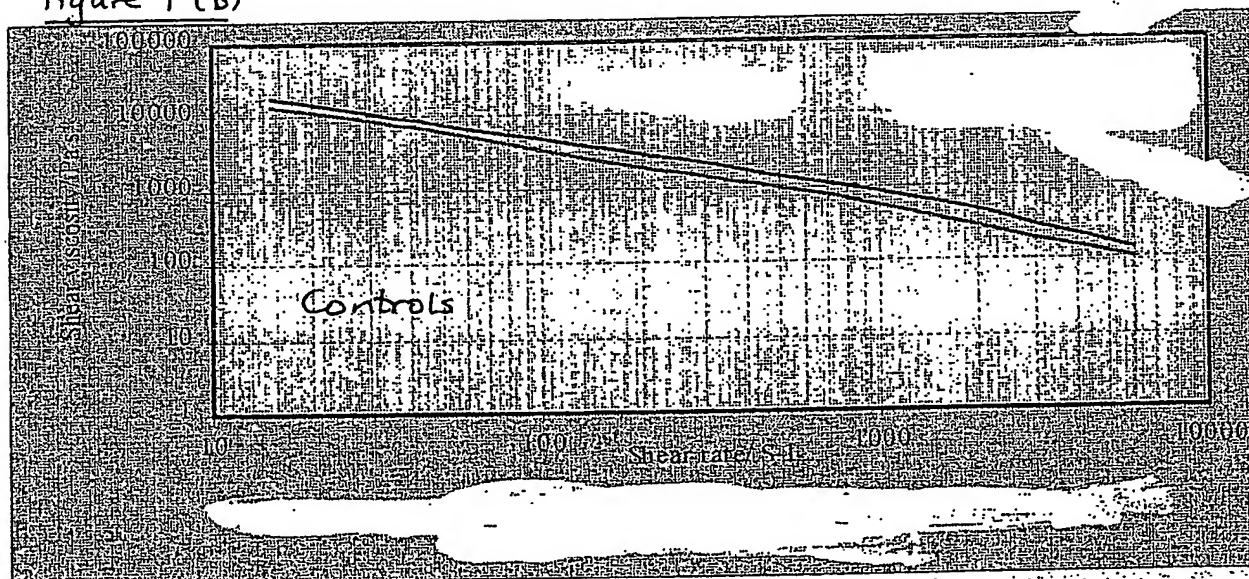
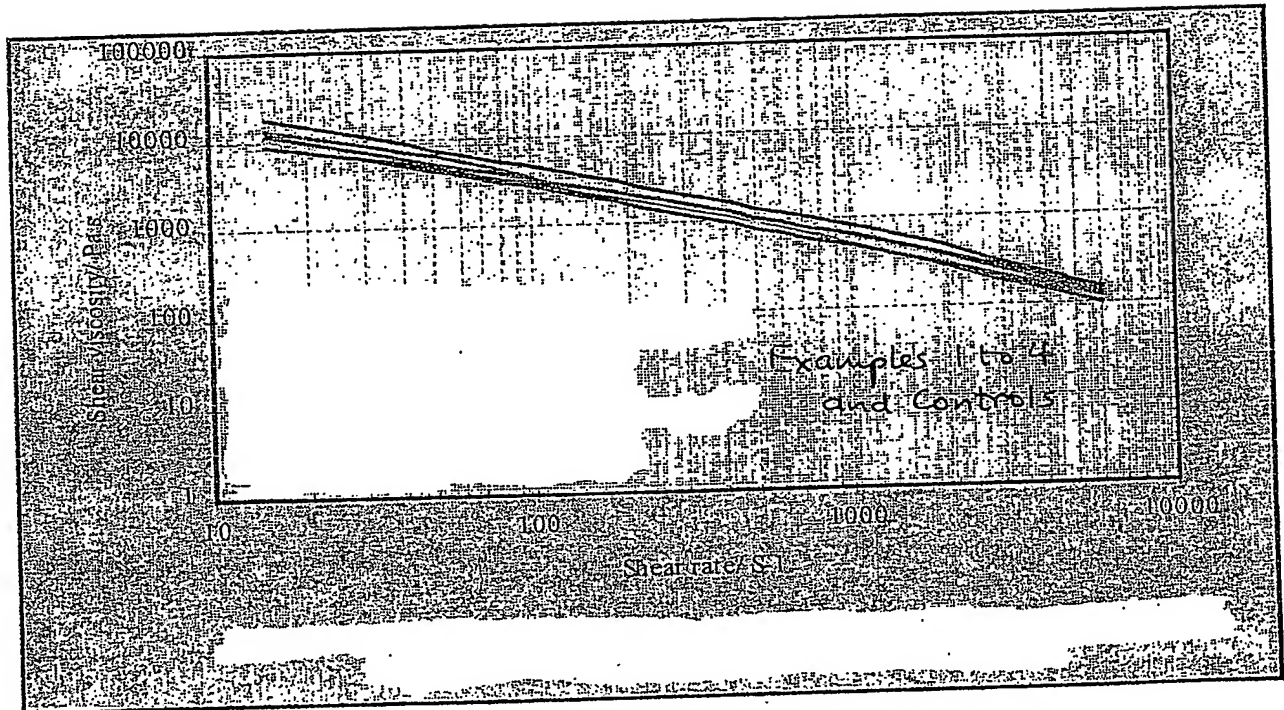


Figure 2



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